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PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT
(PCT Article 36 and Rule 70)

25 MAY 2005

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Applicant's or agent's file reference P200316	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/EP 02/14463	International filing date (day/month/year) 18.12.2002	Priority date (day/month/year) 18.12.2002
International Patent Classification (IPC) or both national classification and IPC C08F110/02		
Applicant BOREALIS TECHNOLOGY OY		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 9 sheets, including this cover sheet.
 - This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 5 sheets.

3. This report contains indications relating to the following items:
 - I Basis of the opinion
 - II Priority
 - III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
 - IV Lack of unity of invention
 - V Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
 - VI Certain documents cited
 - VII Certain defects in the international application
 - VIII Certain observations on the international application

Date of submission of the demand 14.07.2004	Date of completion of this report 04.04.2005
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I. Basis of the report

1. With regard to the elements of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, Pages

1-27 as originally filed

Claims, Numbers

1-25 received on 10.03.2005 with letter of 09.03.2005

2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- the language of publication of the international application (under Rule 48.3(b)).
- the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- contained in the international application in written form.
- filed together with the international application in computer readable form.
- furnished subsequently to this Authority in written form.
- furnished subsequently to this Authority in computer readable form.
- The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- the description, pages:
- the claims, Nos.:
- the drawings, sheets:

5. This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

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III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:
 - the entire international application,
 - claims Nos. 1-28 (in part)

because:

 - the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (specify):
 - the description, claims or drawings (*indicate particular elements below*) or said claims Nos. are so unclear that no meaningful opinion could be formed (*specify*):
 - the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.
 - no international search report has been established for the said claims Nos.
2. A meaningful international preliminary examination cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:
 - the written form has not been furnished or does not comply with the Standard.
 - the computer readable form has not been furnished or does not comply with the Standard.

IV. Lack of unity of invention

1. In response to the invitation to restrict or pay additional fees, the applicant has:
 - restricted the claims.
 - paid additional fees.
 - paid additional fees under protest.
 - neither restricted nor paid additional fees.
2. This Authority found that the requirement of unity of invention is not complied with and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.
3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is
 - complied with.
 - not complied with for the following reasons:
4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:
 - all parts.

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the parts relating to claims Nos. 1-28 (in part) .

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims 9,13,14.
	No:	Claims 1-8, 10-12,15-25
Inventive step (IS)	Yes:	Claims
	No:	Claims 1-25.
Industrial applicability (IA)	Yes:	Claims 1-25.
	No:	Claims

2. Citations and explanations

see separate sheet

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Re Item I

Basis of the opinion

The amendments are allowable.

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The Applicant paid for all search fees but has submitted no response to form PCT/IPEA/405. The subject matter pertaining to invention group 1 (see PCT/IPEA/405) is therefore examined here:

The subject matter of claims 1-25 insofar as it relates to halogen-containing compounds of group 13 of the Periodic Table.

The subject matter pertaining to invention groups 2 , ie the subject matter of claims 1-25 insofar as it relates to halogen-containing compounds of group 14 of the Periodic Table, is not examined here (see PCT/IPEA/405).

Re Item IV

Lack of unity of invention

The present application is found to contravene the requirements of unity of invention according to Art. 3(4)(iii) PCT, Art. 17(3)(a) PCT and Rule 13 PCT for the following reasons: the concept underlying the claims of the present application is the synthesis and use of a support comprising a magnesium hydrocarbyl oxy compound and a halogen containing compound in the production of olefin polymers. However, this concept is well known (see US4496660, example 22 and claims 13 and 15).

According to the present application, the problem relating to the production of this polymer can be solved in several ways as grouped below. Both these groups are linked by the afore-mentioned concept as same or corresponding feature. However, in the light of US4496660, there is no single general inventive concept (Rule 13.1 PCT) and no demonstrated same or corresponding special technical feature (Rule 13.2, PCT) linking these groups:

1. The subject matter of claims 1-25 insofar as it relates to halogen-containing compounds of group 13 of the Periodic Table.
2. The subject matter of claims 1-25 insofar as it relates to halogen-containing

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compounds of group 14 of the Periodic Table.

Re Item V

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

The following documents (D1,D3-D4) will be referred to (see the ISR for additional relevant passages):

D1: US-A-4496660

D3: WO-A-9955741

D4: WO0155230

With regard to the present examples, the present application essentially discloses washing of Al/Mg support prior to contact with group IV metal. 2.28 mol EtAlCl₂ added to octadecafluoroctane (PFO) with stirring to form a emulsion, then (2.24 octMgbu + 2 2-ethyl-hexanol/heptane, 45°C) added to form a suspension. Due to the high density of PFO, the product floats. Liquids are siphoned off and washed with stirring once or twice with heptane (this appears to form the basis of the invention as expressed in the claims, along with catalyst loading). More heptane is then added and 1.12 mol TiCl₄ added with mixing. Heating to 70°C for 45 min follows. Settling followed by removal of supernatant and washing 2X with heptane is undertaken. The product is used with Et₃Al cocatalyst in the homopolymerisation of ethylene. Present example 4 of invention does without the PFO emulsion. Results: Al/Mg : 0.15-0.95:1; Al/Ti : 0.17-0.52:1. Example 4 is compared with a comparative example where no washing of the Mg/Al support takes place: the resulting polymer shows 6.2% of "very fine material" and additionally > 50% fines agglomerated into aggregates, 15% of which had an irregular shape.

1. D1 US4496660 describes the following preparation in the example 22 : 50 mol equiv Bu₂Mg + 275 mol equiv PrOH + 25 mol equiv iBu₃Al + 50 mol equiv SiCl₄ (a group 14 element halide). The product is decanted and washed in hexane. SiCl₄ is capable of halogenating iBu₃Al, a reaction far more plausible than that of halogenating MgOR₂ to MgCl₂. Nevertheless the precise nature of the chemistry involved in this system such as the oxidation state of the Ti metal (vide infra) in the light of any reduction that may take place is unknown but it is in any case the responsibility of the Applicant to prove any differences that may result so long as they are expressed in the present claims. This goes for the order of addition also. The mol ratio of group 14 halide employed to Mg in the process is 1, although the ratio of (group 13+14 elements) : Mg in the product is 1.5, of which the proportion of group 13 and 14 compounds therein which are

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halogenated in the final product is unknown, ie the Applicant has not proved that the amount of group 13 compound comprising halogen falls outside that of the present claims. Then 200 mol equiv of TiCl₄ is added. Thus the TiCl₄: Mg mole ratio is 4.0. The so-obtained catalyst is employed in ethylene polymerisation in the presence of iBu₃Al. Low reactor fouling results. (see eg 22 and claims 13 and 21). Recovery of the support prior to its use as a carrier material is not expressed in the present claims and the presence of other elements is not excluded, thus "should be" (p.5, I.9), does not amount to "is". Hence, in the light of this and Box VIII.5 below, claims 1-8, 10-12, 15-19, 21-23 and 25 are not novel and no analysis of inventivity is required from here on for these claims (note, however that an activity of 191 kg PE/g Ti refers to D3 not D1 and in any case the comparison has not been shown to have taken case under otherwise identical conditions).

2. D3 WO9955741 describes in the examples the following preparation: Cat A: 0.027 mol Bu₂Mg /heptane + 0.014 mol diisoamyl ether (ED) + (chlorinating agent: 0.05 mol Et₂AlCl + 0.1 mol ROH) added with stirring to precipitate a "MgCl₂ derivative". The ROH employed is 2-ethyl-hexanol. The so-produced support is decanted and washed 4 times with hexane. Then 0.06 mol TiCl₄ added. The resulting catalyst loadings are as follows: cat A: Al/Mg: 0.30 mol; Al/Ti: 0.14 mol; cat B: Al/Mg : 0.125 mol; Al/Ti: 0.22 mol. These loadings are very similar to those resulting from the processes of the present examples. The composition is employed in ethylene polymerisation in the presence of iBu₃Al, and results in polymers with low amounts of fines, narrow particle distribution and high bulk density and regular shape. Many types of ROH, including 2-ethyl-1-hexanol are used in the preparation (see pages 6-9, eg1 and table 1; claims 1 and 13). Neither the nature of the hydrocarbon group of the chlorinating agent nor recovery of the support prior to its use as a carrier material is expressed in the present independent claims (so additionally as a consequence, the catalyst loadings do not have to be viewed in the light of the latter). Again, as pointed out in point 1 above, the nature of the chemical reactions that take place are also not expressed in the present claims, so that any speculation about them (which, moreover, remains speculation and not proven fact), cannot be used to establish the novelty of the present claims over D3. Hence claims 15-25 are not novel (see also Box VIII.5).

3. (i) The subject-matter of claims 15 and 16 differs from D3 in that alcohols such as 2-ethyl-1-hexanol are combined with MgR₂ to form hydrocarbyl oxy compounds (feature 1). The technical effect of this feature not been demonstrated, especially as the resultant chemical composition of the compositions of D3 are so similar to those of the present application. Therefore the objective problem can be formulated as to provide

alternative catalysts for olefin polymerisation. The solution proposed in claims 15 and 16 differs of the present application cannot be considered as involving an inventive step because the skilled person would regard it as a normal option to introduce the magnesium reagent as hydrocarbyloxy compound, as shown, for example, in D1 above. Hence claims 13 and 14 are not inventive.

(ii) Likewise, it is trivial to replace Et₂AlCl chlorinating agent with the group 13 halide EtAlCl₂ of the present application, hence claim 9 is not inventive.

4. D4 WO0155230 describes MgOR₂ prepared from MgR₂ and 2-ethyl-hexanol, combined with EtAlCl₂, precipitation being prevented by adding toluene (see eg 1). This composition is isolated without washing and later combined with TiCl₄ to form an olefin polymerisation catalyst (see egs 1-6). Since the order of addition of reagents is not expressed in the present independent claims, in the light of Box VIII.5 claims 1,2,8 and 25 are not novel.

Re Item VIII

Certain observations on the international application

The following objections are made under Art. 6 (PCT):

1. Claims 1, 3 and 4 : (i) "in the obtained reaction product" describes "a results to be achieved" (PCT GL Ch.-III,4.7).

(ii) Claim 1: "to adjust..." describes a "result to be achieved" (PCT GL Ch.-III,4.7).

(iii) These claims should have been defined in terms of all the essential features necessary to carry out the invention. Regardless any treachings in the description, the claims have to be clear in their own right: how would one know what separation and/or washing technique is required to effectively modulate the molar ratios mentioned in these claims over the entire scope of the claims? The amendment to claim 1 thus further exacerbates this serious objection.

2. (i) Claim 11, taken in conjunction with claim 1, describes a process for producing a support which employs a solution of a magnesium hydrocarbyl oxy compound which is itself produced according to claim 13. The magnesium hydrocarbyl oxy compound referred to in claim 1 is a product which may result from the reaction mixture of claim, but cannot be equated with said reaction mixture: the two are distinct entities. Moreover, it cannot be ascertained that the resultant "magnesium hydrocarbyl oxy compound" was prepared by means of a reaction between an "alcohol" and a "magnesium alkyl". It could equally have been prepared from a reaction between a

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magnesium aryl and an alcohol. If it is intended that the "magnesium hydrocarbyloxy compound" be protected as part of the overall claimed process, then the claim should have been redrafted accordingly: if the magnesium hydrocarbyl oxy compound is to be used as prepared in situ, then claim 1 should have been reformulated entirely as a process claim in order to reflect this. As it stands, claim 1 is a process claim which makes use of magnesium products themselves prepared by other processes mentioned in claim 11 (ie claim 1 in conjunction with claim 11 contains a nested product by process claim). This applies to claims 12-14 also (see similarly point 2 above)..

3. Claims 15-17,19,20 are "product-by-process" claims. It cannot be ascertained that the catalyst support product was prepared in this way: this is not the only way that these supports could have been prepared and once the synthons have been consumed in the preparation, there will no longer be any evidence of their existence in order to characterise the products. These claims should have been redrafted as a pure process claim comprising the preparative steps of producing said supports: the claims must be clear in their own right.

4. Claims 21-25 describe processes employing a solid catalyst which is itself produced by a process. It cannot be ascertained that such a solid catalyst was produced in this way (see similarly points 2 and 3 above for further clarification).

5. The subject matter related to the above-mentioned objections 1-4 will be ignored with respect to any considerations that might apply under Box V, since being unclear, it cannot be considered limiting for the scope of the claims.

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Borealis Polymers Oy

CLAIMS:

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1. A process for producing a particulate support for an olefin polymerisation catalyst, wherein a solution of a magnesium compound is contacted with a solution of an element of Group 13 or 14 of the Periodic Table (IUPAC) to obtain a solid reaction product, characterised in that the solid reaction product is formed by

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(i) contacting (a) a solution of a magnesium hydrocarbyloxy compound with (b) a solution of a halogen-containing compound of an element of Group 13 or 14 of the Periodic Table (IUPAC); and
(ii) recovering the solidified reaction product from the reaction mixture by separating the solid product from the liquid reaction medium and/or by washing the product to adjust the molar ratio of the element of Group 13 or 14 of the Periodic Table to magnesium in the obtained reaction product material to a value of at least 0.3, preferably of at least 0.4.

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2. A process of claim 1, wherein the magnesium hydrocarbyloxy compound is of formula (I):
 $Mg(OR_1)_{2-n}(R_1)_nX_x$ (I), wherein each R_1 independently represents a C_{1-20} hydrocarbyl group; X is a halogen; $0 \leq n < 2$ and may or may not be an integer; $x < 2$ and may or may not be an integer; the sum of $(2-n)$, n and x is 2.

3. A process according to claim 1, wherein the molar ratio is adjusted to $0.4 \leq$ (halogen-containing compound of an element of Group 13 or 14):Mg ≤ 1.1 , and preferably to $0.6 \leq$ (halogen-containing compound of an element of Group 13 or 14):Mg ≤ 0.99 .
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4. A process according to claim 1, 2 or 3, wherein said molar ratio is adjusted by washing the obtained reaction product with a wash solution.
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5. A process according to any one of claims 1 to 4, wherein the wash solution is an inert hydrocarbon selected from a linear or branched aliphatic, alicyclic or aromatic C₅₋₂₀ hydrocarbon or any mixtures thereof,
15 and, optionally, the washing step is carried out in a temperature between 40 to 80 °C.
15
6. A process according to any one of claims 1 to 5, wherein (a) the solution of a magnesium hydrocarbyloxy compound is added to (b) a solution of a halogen-containing compound of Group 13 or 14 of the Periodic Table to obtain the solid reaction product.
20
7. A process according to any one of claims 1 to 6, wherein the halogen-containing compound of Group 13 or 14 of the Periodic Table is a chlorine-containing compound of Group 13 of the Periodic Table.
25
8. A process according to any one of claims 1 to 7, wherein the chlorine-containing compound of Group 13 of the Periodic Table is a compound of formula Al(R₁)_xX_{3-x} (II), wherein each R₁ independently represents a C₁₋₂₀ hydrocarbyl group; X is chloride and $0 \leq x < 3$.
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9. A process according to any one of claim 1 to 8, wherein the compound of formula (II) is ethylaluminium dichloride.

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10. A process according to any one of claims 1 to 9, wherein the magnesium hydrocarbyloxy compound is of formula (I): $Mg(OR_1)_{2-n}(R_1)_nX_x$ (I), wherein $0 \leq n < 2$ and may or may not be an integer; each X and R_1 are independently as defined in claim 2; and x is 0.

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11. A process according to any one of claims 1 to 10, wherein the solution of the magnesium hydrocarbyl oxy compound (I) is a reaction mixture prepared by

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contacting in an inert hydrocarbon solvent or any mixtures thereof a magnesium alkyl of formula $Mg(R_1)_2$ (III), wherein each R_1 is independently as defined in claim 2, with an alcohol of formula R_1OH , wherein R_1 is as defined in claim 2, preferably a C_{3-15} cycloalkyl or branched or unbranched C_{3-15} alkyl.

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12. A process according to claim 11, wherein the magnesium alkyl compound (III) is butyloctylmagnesium.

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13. A process according to claim 11 or 12, wherein the alcohol R_1OH is 2-ethyl-1-hexanol.

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14. A process according to any one of claims 12 to 13, wherein butyloctylmagnesium in an inert hydrocarbon solvent or any mixtures thereof is contacted with 2-ethyl-1-hexanol and the obtained solution is added to a solution of ethylaluminium dichloride in an inert

hydrocarbon solvent or any mixtures thereof to form a solid reaction product.

15. A solid catalyst support for an olefin polymerisation catalyst obtainable by a method of any one of claims 1 to 14.

16. A solid catalyst support according to claim 15, wherein the molar ratio of the element of Group 13 or 14 of the Periodic Table (IUPAC) to magnesium in said support is 10 of ≥ 0.3 , preferably ≥ 0.4 .

17. A solid catalyst support for an olefin polymerisation catalyst comprising a separated and/or washed solid reaction product of (a) a magnesium hydrocarbyloxy compound and (b) a halogen-containing compound of an element of Group 13 or 14 of the Periodic Table (IUPAC, the molar ratio of the element of Group 13 or 14 to magnesium in said support being of ≥ 0.3 , preferably $\geq 20 0.4$; and, optionally, of an electron donor.

18. A solid catalyst support according to claim 17, which comprises a separated and/or washed solid reaction product of (a) a reaction mixture of a solution of magnesium alkyl of formula $Mg(R_1)_2$ (III), wherein each R_1 is independently as defined in claim 2, with an alcohol of formula R_1OH , wherein R_1 is as defined in 25 claim 11, in an inert hydrocarbon solvent or any mixtures thereof; and (b) a solution of formula $Al(R_1)_xX_{3-x}$ wherein each R_1 and X and x are as defined in claim 10, in an inert hydrocarbon solvent or any 30 mixtures thereof.

19. A solid catalyst support according to any one of claims 17 to 18, wherein the molar ratio of Al:Mg in said support is ≥ 0.4 , preferably $0.6 \leq \text{Al:Mg} \leq 0.99$.

5 20. A solid support according to claim 18 or 19, wherein in the alcohol of formula R_1OH , R_1 is a C_{3-15} cycloalkyl or branched or unbranched C_{3-15} alkyl.

10 21. A process for producing a Ziegler-Natta catalyst component for olefin polymerisation comprising treating, in an inert solvent, a solid catalyst support according to any one of claims 15 to 20, or prepared according to a method of any one of claims 1 to 14, with a transition metal compound of Group 3 to 10 of the Periodic Table (IUPAC), and, optionally, with an electron donor, and then, optionally, recovering the catalyst component.

15 22. A process according to claim 21, wherein the transition metal compound is a tetravalent titanium compound.

20 23. A process according to claim 22, wherein the transition metal compound is titanium tetrachloride.

25 24. A process according to claim 23, wherein $TiCl_4$ is used in a molar ratio of 1 - 0.5 mol to one mol of Mg present in the support.

30 25. A process for (co)polymerising an olefin using the catalyst component produced according to any one of claims 21 to 24.

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